PERTURBATIVE IMPLEMENTATION OF THE FURRY PICTURE

MATTHIAS HUBER AND EDGARDO STOCKMEYER

ABSTRACT. Recently the block-diagonalization of Dirac-operators was investigated from a mathematical point of view in the one-particle case [13]. We extend this result to the N-particle case. This leads to a perturbative realization of the Furry picture in the N-particle two-spinor space.

1. INTRODUCTION

The idea of block-diagonalizing the Dirac operator, i.e., decoupling electronic and positronic states in such a way that the upper components of a 4-spinor correspond to electronic and the lower components to positronic states, goes back to Foldy and Wouthuysen [4]. They succeeded in decoupling the free Dirac operator and also addressed the case with interaction in the non-relativistic limit. Unfortunately, their expansion does not converge (see Thaller [15, chapter 6] and references therein).

The reason is that the correct parameter for the expansion, in order to have convergence of the spectrum, is not the inverse velocity of light but the coupling constant of the external potential. A perturbative and iterative method to accomplish this is due to Douglas and Kroll [3] and was corrected by Jansen and Heß [5]. The method is very attractive for numerical calculations because the resulting Hamiltonians operate on two-spinors and has been successfully used for calculations in relativistic quantum chemistry in the last twenty years (see [1, 5, 7, 16, 11, 12] and references therein).

From a mathematical point of view, the one-particle case was investigated recently by Siedentop and Stockmeyer [14] (see also [13]). They proved, under suitable conditions on the potential, that there exists a family U_γ of unitary operators, analytic in the coupling constant γ, that exactly decouples electronic and positronic states. Moreover, it was shown that the block-diagonalized Dirac operator developed in power series in γ coincides, at least formally in the first orders, with the operators resulting from the method of Douglas, Kroll and Heß. Moreover, they proved that the spectra of the truncated expansions converge to the spectra of the original operator.

The aim of this work is to extend their results to the N-particle Coulomb-Dirac Hamiltonian. We consider the N-particle Coulomb-Dirac operator in the Furry picture, i.e., the operator restricted to the positive spectral subspaces of each one particle operator. Using simple generalizations of the methods used in [14] we prove norm resolvent convergence of the operators resulting from the power expansion of the projected Hamiltonian in powers of the coupling constant. For convenience for

the reader, we give in section 5 the main definitions and results of [14], as far as we need them.

2. DEFINITION OF THE PROBLEM AND NOTATION

The N-particle Hilbert space is denoted by

$$\mathfrak{H}^{(N)} = \mathfrak{H} \otimes \cdots \otimes \mathfrak{H},$$

where $\mathfrak{H} := L^2(\mathbb{R}^3, \mathbb{C}^4)$. An extension of a closable operator A on $\mathfrak{H}$ with domain $\mathcal{D}$ to $\mathfrak{H}^{(N)}$ acting on the j-th component is written as

$$A_j := \mathbf{1} \otimes \cdots \otimes \underbrace{A}_{j\text{-th place}} \otimes \cdots \otimes \mathbf{1},$$

and the n-fold tensor product as

$$\mathcal{A} := A \otimes \cdots \otimes A.$$

If A is essentially self-adjoint on $\mathcal{D}$, then all of the above operators are essentially self-adjoint on $\bigotimes_{j=1}^{N} \mathcal{D}$ (see [9], chapter VIII.10). For technical reasons, we will need the operator $\mathscr{D}_0 := \sum_{j=1}^{N} |D_0|_j$, where D_0 is the free Dirac operator.

The Coulomb-Dirac operator is given formally by

$$H_{CD}^{(N)} := \sum_{j=1}^{N} [D_\gamma]_j + \frac{\gamma}{Z} \sum_{1 \le i < j \le N} W_{ij}, \tag{1}$$

where, using units $\hbar = c = 1$, Z is the atomic number of the nucleus, $\gamma = Z\alpha^2$ is the coupling constant of the one-particle potential, and α is the fine structure constant. $D_\gamma = D_0 + \gamma V$ where D_0 is the free Dirac operator, $V = -1/|\cdot|$ is the Coulomb interaction with the nucleus, and W_{ij} is the interaction between the particle i and j, which is defined by

$$(W_{ij}f)(x_1, \ldots, x_N) := \frac{f(x_1, \ldots, x_i, \ldots, x_j, \ldots, x_N)}{|x_i - x_j|} \tag{2}$$

for $f \in H^1(\mathbb{R}^3)^4 \otimes \cdots \otimes H^1(\mathbb{R}^3)^4$. We set $W_\gamma := \frac{\gamma}{Z} \sum_{1 \le i < j \le N} W_{ij}$. It is well known that the operator in (1) (without the electron-electron interaction W_γ) has the whole real axis as its spectrum.

We consider instead of (1) the Coulomb-Dirac operator in the Furry picture, that is we restrict (1) onto the positive spectral subspaces of each one-particle operator D_γ. The Hilbert space is given by

$$\mathfrak{H}_+^{(N)}(\gamma) = P_+^\gamma \mathfrak{H} \otimes \cdots \otimes P_+^\gamma \mathfrak{H},$$

where $P_+^\gamma := \chi_{[0,\infty)}(D_\gamma)$. The N-particle projection is given by

$$\mathcal{P}_+^\gamma := P_+^\gamma \otimes \cdots \otimes P_+^\gamma. \tag{3}$$

We are interested in $\mathcal{P}_+^\gamma H_{CD}^{(N)} \mathcal{P}_+^\gamma$ in particular, in its realization in the N particle two-spinor space. We recall the definition and properties of the unitary transformation U_γ given in [14] (see equation (22) below) and consider its N-particle version

$$\mathcal{U}_\gamma := U_\gamma \otimes \cdots \otimes U_\gamma. \tag{4}$$

This operator has the property $\mathcal{U}_\gamma \mathcal{P}_+^\gamma \mathcal{U}_\gamma^{-1} = \mathcal{P}_+^0$. Moreover, the N-particle Foldy-Wouthuysen transformation $\mathcal{U}_{\mathrm{FW}}$ fulfills $\mathcal{U}_{\mathrm{FW}} \mathcal{P}_+^0 = \beta_+ \otimes \cdots \otimes \beta_+ \mathcal{U}_{\mathrm{FW}}$ where $\beta_+ := (1+\beta)/2$ is the projection onto the upper two-spinor. Therefore, the formal Hamiltonian in the Furry picture realized in the N-particle two-spinor space is given by

$$H_{\mathrm{diag}}^N = \mathcal{U}_{\mathrm{FW}} \mathcal{U}_\gamma \mathcal{P}_+^\gamma H_{CD}^{(N)} \mathcal{P}_+^\gamma \mathcal{U}_\gamma^{-1} \mathcal{U}_{\mathrm{FW}}^{-1}. \tag{5}$$

Our main result, Theorem 2 below, is that the spectra of the operators resulting from the expansion in γ of (5) converge to the spectrum of the Furry-operator given formally by $\mathcal{P}_+^\gamma H_{CD}^{(N)} \mathcal{P}_+^\gamma$.

3. Main result

The one-particle Dirac operator with Coulomb potential is given by

$$D_\gamma := \boldsymbol{\alpha} \cdot \tfrac{1}{i}\nabla + \beta - \gamma \frac{1}{|\cdot|} \tag{6}$$

acting in the Hilbert space $L^2(\mathbb{R})^4$. For $|\gamma| < \sqrt{3}/2$ the operator D_γ is essentially self-adjoint on $C_0^\infty(\mathbb{R}^3 \setminus \{0\})^4$ and self-adjoint on $H^1(\mathbb{R}^3)^4$. Furthermore, for $\gamma < 1$ the operator has a distinguished self-adjoint extension characterized by $\mathfrak{D}(D_\gamma) \subset H^{1/2}$ (see Thaller [15, Theorem 4.4.]). Moreover, for $|\gamma| < 1$ we have that $|D_\gamma| \geq \sqrt{1-\gamma^2} > 0$.

We define the operator $\tilde{T}_\gamma : \mathfrak{H}_+^{(N)} \to \mathfrak{H}_+^{(N)}$ without electron-electron interaction as

$$\tilde{T}_\gamma = \mathcal{P}_+^\gamma \sum_{i=1}^N [D_\gamma]_i \mathcal{P}_+^\gamma, \tag{7}$$

with domain $\mathfrak{D}(\tilde{T}_\gamma) = \mathcal{P}_+^\gamma H^1(\mathbb{R}^3, \mathbb{C}^4)^{\otimes N}$. For $|\gamma| < \sqrt{3}/2$ the operator $\tilde{T}_\gamma$ is essentially self-adjoint ([9, Theorem VIII.33]). We denote the closure of $\tilde{T}_\gamma$ by T_γ and its form domain by $\mathfrak{Q}(T_\gamma)$.

Define the quadratic form $q(f,g) := (f, T_\gamma g) + (f, W_\gamma g)$ for $f, g \in \mathfrak{Q}(T_\gamma)$. We have the following:

Theorem 1. *Let $0 \leq \gamma < \sqrt{3}/2$. There exists a unique self-adjoint operator $H_{\gamma,+} : \mathfrak{H}_+^{(N)}(\gamma) \to \mathfrak{H}_+^{(N)}(\gamma)$ with $\mathfrak{Q}(H_{\gamma,+}) = \mathfrak{Q}(T_\gamma) = \mathcal{P}_+^\gamma H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$ where $\mathcal{P}_+^\gamma (H^1(\mathbb{R}^3)^4)^{\otimes N}$ is a form core for $H_{\gamma,+}$, such that for all $f, g \in \mathfrak{Q}(H_{\gamma,+})$*

$$q(f,g) = (f, H_{\gamma,+}g).$$

This self-adjoint extension is the Friedrich extension of the symmetric operator $T_\gamma + \mathcal{P}_+^\gamma W_\gamma \mathcal{P}_+^\gamma$ defined on $\mathcal{P}_+^\gamma \bigotimes_{j=1}^N H^1(\mathbb{R}^3)^4$.

Proof. Step 1: Well-definedness of q. We have by Lemma 6

$$(f, |D_0|_i f) \leq \frac{1}{d_\gamma}(f, |D_\gamma|_i f) \leq \frac{1}{d_\gamma}(f, T_\gamma f). \tag{8}$$

This inequality together with Lemma 1 implies the well-definedness of q.

Step 2: Definition of $H_{\gamma,+}$. We mimick the proof of the KLMN-theorem, using the notation of of Reed-Simon [10, Theorem X.17]. Pick $f \in \mathfrak{Q}(T_\gamma)$. We start by proving that $(f, W_\gamma f) \leq c(f, T_\gamma f)$ for some $c > 0$. Since for $f \in \mathfrak{H}_+^{(N)}$, we have

$$\begin{aligned}
(T_\gamma^{-1/2}f, & W_\gamma T_\gamma^{-1/2}f) = \\
&= \frac{\gamma}{Z} \sum_{1 \leq i < j \leq N} (T_\gamma^{-1/2}f, |D_0|_i^{1/2}|D_0|_i^{-1/2}W_{ij}|D_0|_i^{-1/2}|D_0|_i^{1/2}T_\gamma^{-1/2}f) \\
&\leq \frac{\gamma\pi}{2Z} \sum_{1 \leq i < j \leq N} (T_\gamma^{-1/2}f, |D_0|_i T_\gamma^{-1/2}f) \leq \frac{\gamma\pi N(N-1)}{4Zd_\gamma}\|f\|^2 =: c\|f\|^2,
\end{aligned} \tag{9}$$

where we used Lemma 1 and equation (8). Therefore,

$$(f, T_\gamma f) + (f, W_\gamma f) \leq (1+c)(f, T_\gamma f) \leq (1+c)(f, (T_\gamma + W_\gamma)f). \tag{10}$$

The latter shows that the norms $\|\cdot\|_{+1,T_\gamma}$ and $\|\cdot\|_{+1,q}$ are equivalent. Thus q is a semi-bounded, closed quadratic form on $\mathfrak{Q}(T_\gamma)$, which therefore defines a self-adjoint operator $H_{\gamma,+}$ with form-domain $\mathfrak{Q}(H_{\gamma,+}) = \mathfrak{Q}(T_\gamma)$.

Step 3: Determination of the form domain. We have the following chain of in-equatlities, where we used Lemma 6 in the second and Hardy's inequality in the third step:

$$d_\gamma \sqrt{-\triangle_{3N} + 1} \le d_\gamma \sum_{i=1}^{N} |D_0|_i \le \sum_{i=1}^{N} |D_\gamma|_i \le$$

$$\sum_{i=1}^{N} (1 + 2\gamma)|D_0|_i \le (1 + 2\gamma)N\sqrt{-\triangle_{3N} + 1} \quad (11)$$

Inequalities (11) imply that $H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$ is complete with respect to the quadratic form $b(f, f) := (\sqrt{\sum_{i=1}^{N} |D_\gamma|_i} f, \sqrt{\sum_{i=1}^{N} |D_\gamma|_i} f)$ for $f \in H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$. Since $\mathcal{P}_+^\gamma$ commutes with $\sum_{i=1}^{N} |D_\gamma|_i$, we have $\mathcal{P}_+^\gamma H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N}) \subset H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$. Moreover, $\mathcal{P}_+^\gamma H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$ is a closed subspace of $H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$ with respect to the norm generated by b. Hence $\mathcal{P}_+^\gamma H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$ is complete with respect to the restriction of b. Since $\tilde{T}_\gamma$ is essentialy self-adjoint the self-adjoint operator associated to the restriction of b is T_γ. Thus, $\mathfrak{Q}(T_\gamma) = \mathcal{P}_+^\gamma H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$.

Since obviously $\mathfrak{D}(H_{\gamma,+}^N) \subset \mathfrak{Q}(H_{\gamma,+}^N) = \mathcal{P}_+^\gamma H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$, we have that $H_{\gamma,+}^N$ is the Friedrich extension of the symmetric operator $\tilde{T}_\gamma + \mathcal{P}_+^\gamma W_\gamma \mathcal{P}_+^\gamma$ defined on $\mathcal{P}_+^\gamma \bigotimes_{j=1}^{N} H^1(\mathbb{R}^3)^4$. $\square$

We now turn to the operator $\mathcal{U}_\gamma := U_\gamma \otimes \cdots \otimes U_\gamma$ which is a unitary mapping $\mathcal{U}_\gamma : \mathfrak{H}_+^{(N)}(\gamma) \to \mathfrak{H}_+^{(N)}(0)$. We define the operator $\tilde{H}_{\gamma,+} : \mathfrak{H}_{+,0}^{(N)} \to \mathfrak{H}_{+,0}^{(N)}$ as $\tilde{H}_{\gamma,+} := \mathcal{U}_\gamma H_{\gamma,+} \mathcal{U}_\gamma^{-1}$, where $\mathfrak{H}_{+,0}^{(N)} := \mathfrak{H}_+^{(N)}(0)$. Analogously, interpreting the Foldy-Wouthuysen transformation in a natural way as a mapping $\mathcal{U}_{\text{FW}} : \mathfrak{H}_{+,0}^{(N)} \to L^2(\mathbb{R}^3, \mathbb{C}^2)^{\otimes N} = L^2(\mathbb{R}^{3N}, \mathbb{C}^{2^N})$, we define $H_{\gamma,+}^{\text{diag}} : L^2(\mathbb{R}^{3N}, \mathbb{C}^{2^N}) \to L^2(\mathbb{R}^{3N}, \mathbb{C}^{2^N})$ as the operator $H_{\gamma,+}^{\text{diag}} = \mathcal{U}_{\text{FW}} \tilde{H}_{\gamma,+} \mathcal{U}_{\text{FW}}^{-1}$. In this way, $H_{\gamma,+}^{\text{diag}}$ can be seen as the block-diagonalization of $\tilde{H}_{\gamma,+}$. By Theorem 1, $\tilde{H}_{\gamma,+}$ and $H_{\gamma,+}^{\text{diag}}$ are self-adjoint operators with form domain $\mathcal{U}_\gamma \mathfrak{Q}(T_\gamma)$ and $\mathcal{U}_{\text{FW}} \mathcal{U}_\gamma \mathfrak{Q}(T_\gamma)$ respectively. Actually $\mathcal{U}_\gamma \mathfrak{Q}(T_\gamma) = \mathcal{P}_+^0 H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{4^N})$, since $\mathscr{D}_0^{1/2} \mathcal{U}_\gamma \mathscr{D}_0^{-1/2}$ is a bounded operator (see proof of Lemma 3). Since $\mathcal{U}_{\text{FW}}$ commutes with $\mathscr{D}_0$, we get $\mathcal{U}_{\text{FW}} \mathcal{U}_\gamma \mathfrak{Q}(T_\gamma) = H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{2^N})$.

We denote by $\tilde{h}_{\gamma,+}^k$ the formal Taylor expansion of $H_{\gamma,+}^{\text{diag}}$ up to the power γ^k inclusive, acting on $\mathcal{C} := \mathcal{U}_{\text{FW}} \mathcal{U}_\gamma \mathcal{P}_+(\gamma) H^1(\mathbb{R}^3, \mathbb{C}^4)^{\otimes N}$ which is a form core for $H_{\gamma,+}^{\text{diag}}$. We set $R_\gamma^k = H_{\gamma,+}^{\text{diag}} - \tilde{h}_{\gamma,+}^k$.

The main result of this letter is:

Theorem 2. *There exists a $\gamma_c > 0$ such that for $0 \le \gamma < \gamma_c$ the operators $\tilde{h}_{\gamma,+}^k$ admit a distinguished self-adjoint extension $h_{\gamma,+}^k$ for k big enough with the property that $\mathfrak{D}(h_{\gamma,+}^k) \subset \mathfrak{Q}(H_{\gamma,+}^{\text{diag}}) = H^{1/2}(\mathbb{R}^{3N}, \mathbb{C}^{2^N})$. Moreover $h_{\gamma,+}^k \to H_{\gamma,+}^{\text{diag}}$ as $k \to \infty$ in the sense of norm resolvent convergence.*

Proof. According to Kato [6, Theorem VI.3.11 and Corollary VI.3.12] it is enough to prove there exist a sequence a_k with $a_k \to 0$ as $k \to \infty$, such that for any $f \in \mathcal{C}$

$$(f, R_\gamma^k f) \le a_k(f, H_{\gamma,+}^{\text{diag}} f). \quad (12)$$

This is equivalent to

$$(f, H_{\gamma,+}^{-1/2} \mathcal{U}_\gamma^{-1} \mathcal{U}_{\text{FW}}^{-1} R_\gamma^k \mathcal{U}_{\text{FW}} \mathcal{U}_\gamma H_{\gamma,+}^{-1/2} f) \to 0 \quad (13)$$

for $f \in \mathcal{P}_+(\gamma) H^1(\mathbb{R}^3, \mathbb{C}^4)^{\otimes N}$. Then

$$
\begin{aligned}
(f, &H_{\gamma,+}^{-1/2} \mathcal{U}_\gamma^{-1} \mathcal{U}_{\text{FW}}^{-1} R_\gamma^k \mathcal{U}_{\text{FW}} \mathcal{U}_\gamma H_{\gamma,+}^{-1/2} f) \\
&\leq \|\mathscr{D}_0^{-1/2} R_\gamma^k \mathscr{D}_0^{-1/2}\| \, \|\mathscr{D}_0^{1/2} \mathcal{U}_\gamma \mathscr{D}_0^{-1/2}\|^2 \, \|\mathscr{D}_0^{1/2} H_{\gamma,+}^{-1/2} f\|^2,
\end{aligned}
\tag{14}
$$

the last term goes to zero due to lemmas 2, 3 and 4 below. We used in the last step that U_{FW} commutes with $|D_0|$ and therefore $\mathcal{U}_{FW}$ with $\mathscr{D}_0$ and any function of it. $\qquad\square$

Remark 1. *The unitary transform U_γ is not unique. If we take the choice given in [14] (22) in the appendix, we know that $\gamma_c \geq 0,3775$ which corresponds to the critical atomic number $Z = 52$.*

4. Auxiliary lemmas

The following bound is Kato's inequality.

Lemma 1. *Let $f \in H^{1/2}(\mathbb{R}^3 N, \mathbb{C}^{4^N})$. Then*

$$
(f, W_{ij} f) \leq \frac{\pi}{2}(f, |D_0|_j f) \text{ for } i \neq j
\tag{15}
$$

Proof. Pick $f \in \bigotimes_{j=1}^N H^1(\mathbb{R}^3)^4$ arbitrarily.

$$
(f, W_{ij} f) = \int dx_1 \cdots dx_N \overline{f(x_1, \ldots, x_i, \ldots, x_N)} \frac{1}{|x_i - x_j|} f(x_1, \ldots, x_i, \ldots, x_N) =
$$

$$
\int dx_1 \cdots dx_N \overline{f(x_1, \ldots, x_i + x_j, \ldots, x_N)} \frac{1}{|x_i|} f(x_1, \ldots, x_i + x_j, \ldots, x_N) \leq
$$

$$
\frac{\pi}{2} \int dx_1 \cdots dx_N \overline{f(x_1, \ldots, x_i + x_j, \ldots, x_N)} |D_0|_i f(x_1, \ldots, x_i + x_j, \ldots, x_N) \leq
$$

$$
\frac{\pi}{2}(f, |D_0|_i f), \quad (16)
$$

where we used Kato's inequality. This inequality extends by continuity to $f \in H^{1/2}(\mathbb{R}^3 N, \mathbb{C}^{4^N})$. $\qquad\square$

Lemma 2. *For $|\gamma| < \gamma_c$ the operator $\mathscr{D}_0^{-1/2} H_{\gamma,+}^{\text{diag}} \mathscr{D}_0^{-1/2}$ is bounded and real analytic around zero. In particular $\|\mathscr{D}_0^{-1/2} R_\gamma^k \mathscr{D}_0^{-1/2}\| \to 0$.*

Proof. First note that $|D_0|_i^{1/2} \mathscr{D}_0^{-1/2}$ is bounded. To prove this, we take an arbitrary $f \in \bigotimes_{j=1}^N H^1(\mathbb{R}^3)^4$ and note that

$$
\||D_0|_i^{1/2} f\|^2 = (f, |D_0|_1 f) \leq (f, |D_0|_1 f) + \cdots + (f, |D_0|_N f) = \|\mathscr{D}_0^{1/2} f\|^2, \quad (17)
$$

the claim follows by density of $\bigotimes_{j=1}^N H^1(\mathbb{R}^3)^4$ in $\mathfrak{H}^{(N)}$.

According to Lemma 5, the operator $|D_0|^{-1/2} U_\gamma D_\gamma U_\gamma^* |D_0|^{-1/2}$ is bounded and analytic in γ for $|\gamma| < \gamma_c$, and so is $[(|D_0|^{-1/2} U_\gamma D_\gamma U_\gamma^* |D_0|^{-1/2})]_m$ for $m = 1, \ldots, N$. In order to prove the claim it suffices to show the analyticity of each summand of $\tilde{H}_{\gamma,+}$.

One-particle terms: The analyticity follows immediately from

$$
\begin{aligned}
\mathscr{D}_0^{-1/2} [U_\gamma P_+^\gamma D_\gamma P_+^\gamma U_\gamma^{-1}]_m \mathscr{D}_0^{-1/2} = \mathcal{P}_+^0 \mathscr{D}_0^{-1/2} [|D_0|^{1/2}]_m \\
\times [|D_0|^{-1/2} U_\gamma D_\gamma U_\gamma^* |D_0|^{-1/2}]_m |D_0|_m^{1/2} \mathscr{D}_0^{-1/2} \mathcal{P}_+^0. \quad (18)
\end{aligned}
$$

Interaction terms: The operator

$$
|D_0|_m^{-1/2} \mathcal{U}_\gamma |D_0|_m^{1/2} = U_\gamma \otimes \cdots \otimes |D_0|^{-1/2} U_\gamma |D_0|^{1/2} \cdots \otimes U_\gamma
$$

is analytic by Lemma 5, and $|D_0|_m^{-1/2} W_{m,l} |D_0|_m^{-1/2}$ is bounded by Lemma 1. Thus, writing

$$\mathscr{D}_0^{-1/2} \mathcal{U}_\gamma \mathcal{P}_+^\gamma W_{m,l} \mathcal{P}_+^\gamma \mathcal{U}_\gamma^{-1} \mathscr{D}_0^{-1/2} = \mathcal{P}_+^0 \mathscr{D}_0^{-1/2} \mathcal{U}_\gamma W_{m,l} \mathcal{U}_\gamma^{-1} \mathscr{D}_0^{-1/2} \mathcal{P}_+^0 =$$
$$\mathcal{P}_+^0 \mathscr{D}_0^{-1/2} |D_0|_m^{1/2} |D_0|_m^{-1/2} \mathcal{U}_\gamma |D_0|_m^{1/2} |D_0|_m^{-1/2} W_{m,l} |D_0|_m^{-1/2}$$
$$|D_0|_m^{1/2} \mathcal{U}_\gamma^{-1} |D_0|_m^{-1/2} |D_0|_m^{1/2} \mathscr{D}_0^{-1/2} \mathcal{P}_+^0, \quad (19)$$

we have shown the analyticity of the interaction term.

Since $\mathcal{U}_{\mathrm{FW}}$ commutes with $\mathscr{D}_0$ the operator $\mathscr{D}_0^{-1/2} H_{\gamma,+}^{\mathrm{diag}} \mathscr{D}_0^{-1/2}$ is also analytic and therefore has a convergent Taylor expansion with $\|\mathscr{D}_0^{-1/2} R_\gamma^k \mathscr{D}_0^{-1/2}\| \to 0$ as $k \to \infty$. □

Lemma 3. *For $|\gamma| < \gamma_c$ the operator $\mathscr{D}_0^{1/2} \mathcal{U}_\gamma \mathscr{D}_0^{-1/2}$ is bounded on $\mathfrak{H}_0^{(N)}$.*

Proof. We have that $\mathscr{D}_0^{1/2} \le \sum_{i=1}^{N} |D_0|_i^{1/2}$. Note further that by Lemma 5 the operator $|D_0|^{1/2} \mathcal{U}_\gamma |D_0|^{-1/2}$ is bounded, thus

$$\|\mathscr{D}_0^{1/2} \mathcal{U}_\gamma \mathscr{D}_0^{-1/2}\| = \|\mathscr{D}_0^{1/2} (\sum_{i=1}^N |D_0|_i^{1/2})^{-1} \sum_{i=1}^N |D_0|_i^{1/2} \mathcal{U}_\gamma \mathscr{D}_0^{-1/2}\|$$
$$= \|\mathscr{D}_0^{1/2} (\sum_{i=1}^N |D_0|_i^{1/2})^{-1} \sum_{i=1}^N (|D_0|_i^{1/2} \mathcal{U}_\gamma |D_0|_i^{-1/2} |D_0|_i^{1/2} \mathscr{D}_0^{-1/2})\|$$
$$\le \sum_{i=1}^N \||D_0|_i^{1/2} \mathcal{U}_\gamma |D_0|_i^{-1/2} |D_0|_i^{1/2} \mathscr{D}_0^{-1/2})\|$$
$$\le \sum_{i=1}^N \||D_0|_i^{1/2} \mathcal{U}_\gamma |D_0|_i^{-1/2}\| \||D_0|_i^{1/2} \mathscr{D}_0^{-1/2}\|$$

$$(20)$$

is finite. □

Lemma 4. *For $f \in \mathfrak{H}_+^{(N)}(\gamma)$, and $0 \le \gamma < \sqrt{3}/2$ we have the estimate $\|\mathscr{D}_0^{1/2} H_{\gamma,+}^{-1/2} f\| \le 1/\sqrt{d_\gamma} \|f\|$.*

Proof. Pick $f \in \mathcal{P}_+(\gamma) \left(H^1(\mathbb{R}^3)^4\right)^{\otimes N}$. Then using Lemma 6 and $W_\gamma \ge 0$ we get

$$(f, \mathscr{D}_0 f) \le \frac{1}{d_\gamma} (f, \sum_j (P_+^\gamma D_\gamma P_+^\gamma)_j f) \le \frac{1}{d_\gamma} (f, H_{\gamma,+} f). \quad (21)$$

□

5. The one-particle case

Let us define the following operator

$$U_\gamma := (P_+^0 P_+^\gamma + P_-^0 P_-^\gamma)(1 - (P_+^0 - P_+^\gamma))^2)^{-1/2}. \quad (22)$$

Some important properties ([14, Theorem 1, Theorem 2, Lemma 9]) of U_γ are listed in the following lemma. We set $\gamma_c := 0.3775$.

Lemma 5.   *(1) U_γ is analytic in γ and unitary for $|\gamma| < 0.6841$ and fulfills the relation*
$$U_\gamma P_\pm^\gamma = P_\pm^0 U_\gamma.$$
   (2) The operator $|D_0|^{1/2} U_\gamma |D_0|^{-1/2}$ is bounded and analytic in γ for $\gamma < \gamma_c$.

(3) *The operator $|D_0|^{-1/2}U_\gamma D_\gamma U_\gamma^*|D_0|^{-1/2}$ is bounded and analytic in γ for $\gamma < \gamma_c$.*

The following inequality is used in this paper. For $0 \leq \gamma < \frac{\sqrt{3}}{2}$ set $C_\gamma :=$ $\frac{1}{3}\left(\sqrt{4(\gamma)^2+9} - 4\gamma\right)$ and $d_\gamma := \frac{1}{2}(1 + C_\gamma^2 - \sqrt{(1-C_\gamma^2)^2 + 4\gamma^2 C_\gamma^2})$. We have (see Morozov [8] and also Brummelhuis et al [2]):

Lemma 6. *For $0 \leq \gamma < \frac{\sqrt{3}}{2}$ the operator inequality*

$$|D_\gamma|^2 \geq d_\gamma^2 |D_0|^2 \tag{23}$$

holds.

References

[1] Maria Barysz and Andrzej Sadlej. Two-component methods of relativistic quantum chemistry: From the Douglas-Kroll approximation to the exact two-component formalism. *J. Molec. Struc. (Theochem)*, 573:181–200, 2001.

[2] Raymond Brummelhuis, Norbert Röhrl, and Heinz Siedentop. Stability of the relativistic electron-positron field of atoms in Hartree-Fock approximation: heavy elements. *Doc. Math.*, 6:1–9, 2001.

[3] Marvin Douglas and Norman M. Kroll. Quantum electrodynamical corrections to the fine structure of helium. *Annals of Physics*, 82:89–155, 1974.

[4] Leslie L. Foldy and Siegfried A. Wouthuysen. On the Dirac Theory of Spin 1/2 Particles and its Non-Relativistic Limit. *Phys. Rev., II. Ser.*, 78:29–36, 1950.

[5] Georg Jansen and Bernd A. Heß. Revision of the Douglas-Kroll transformation. *Physical Review A*, 39(11):6016–6017, June 1989.

[6] Tosio Kato. *Perturbation Theory for Linear Operators*, volume 132 of *Grundlehren der mathematischen Wissenschaften*. Springer-Verlag, Berlin, 1 edition, 1966.

[7] Werner Kutzelnigg. Relativistic one-electron Hamiltonians 'for electrons only' and the variational treatment of the Dirac equation. *Chemical Physics*, 225(1):203–222, 1997.

[8] Sergey Morozov. Extension of a minimax principle for Coulomb-Dirac operators. Master's thesis, Mathematisches Institut, Ludwig-Maximilians-Universität, Theresienstr. 39, 80333 München, Germany, August 2004.

[9] Michael Reed and Barry Simon. *Methods of modern mathematical physics. I. Functional analysis.* Academic Press, New York, 1972.

[10] Michael Reed and Barry Simon. *Methods of modern mathematical physics. II. Fourier analysis, self-adjointness.* Academic Press [Harcourt Brace Jovanovich Publishers], New York, 1975.

[11] M. Reiher and A. Wolf. Exact decoupling of the Dirac hamiltonian. I. general theory. *J. Chem. Phys*, 121(5):2037–2047, 2004.

[12] M. Reiher and A. Wolf. Exact decoupling of the Dirac hamiltonian. II. the generalized Douglas-Kroll-Hess transformation up to arbitrary order. *J. Chem. Phys*, 121(22):10945–10956, 2004.

[13] Heinz Siedentop and Edgardo Stockmeyer. An analytic Douglas-Kroll-Heß method. *Phys. Lett. A*, 341:473–478, 2005.

[14] Heinz Siedentop and Edgardo Stockmeyer. The Douglas-Kroll-Heß method: convergence and block-diagonalization of Dirac operators. *Ann. Henri Poincaré*, 7(1):45–58, 2006.

[15] Bernd Thaller. *The Dirac Equation.* Texts and Monographs in Physics. Springer-Verlag, Berlin, 1 edition, 1992.

[16] Alexander Wolf, Markus Reiher, and Bernd Hess. The generalized Douglas-Kroll transformation. *J. Chem. Phys.*, 117(20):9215–9226, 2002.

Mathematisches Institut, Ludwig-Maximilians-Universität München, Theresienstrasse 39, 80333 München, Germany

E-mail address: mhuber@math.lmu.de and stock@math.lmu.de